

Na/BETA"-ALUMINA/NaAlCl<sub>4</sub>, Cl<sub>2</sub>/C CIRCULATING CELL

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A study has been made of a high specific energy battery based on a sodium negative electrode and a chlorine positive electrode with molten AlCl<sub>3</sub>-NaCl electrolyte and a solid beta"-alumina separator (open circuit potential = 3.8 V, theoretical energy density = 1740 Wh/kg). The basic performance of a Na/beta"-alumina/NaAlCl<sub>4</sub>, Cl<sub>2</sub>/C circulating cell at 200 °C has been demonstrated. This cell can be started at 150 °C. The use of melting sodium chloroaluminate electrolyte overcomes some of the material problems associated with the high working temperatures of present molten salt systems, such as Na/S and Li-Al/FeS, and retains the advantages of high energy density and relatively efficient electrode processes. Preliminary investigations were conducted on a sodium-chlorine static cell, material compatibility, electrode design, wetting, and theoretical calculations to assure a better chance of success before assembling a Na/Cl<sub>2</sub> circulating cell.

In the sodium-chlorine static cell experiment, an open circuit potential of the sodium-chlorine couple in the range of 3.78-3.81 V was measured. A current density of 10 mA/cm<sup>2</sup> at steady state discharge could be obtained at a 3.0-V cell potential. Causes of the polarization losses were identified as poor wetting of sodium on the surface of beta"-alumina tubes and slow chlorine mass transfer rate to the chlorine electrode.

The materials and corrosion tests reveal that nickel and monel are corrosion resistant in the molten NaAlCl<sub>4</sub>. Carbon cloth and vitreous carbon are the best materials for the chlorine electrode. Grafoil<sup>R</sup> and Teflon<sup>R</sup> were useful as seal and gasket material. In general, the materials used performed well; however, the Monel pump shaft and parts contacting stagnant electrolyte showed a noticeable corrosion rate. The products of corrosion such as CuCl<sub>2</sub> and NiCl<sub>2</sub> affect the performance of the chlorine electrode to some extent.

Diffusivity and solubility of chlorine in NaAlCl<sub>4</sub>, needed in the mathematical models, were measured as functions of temperature. The low solubility and diffusivity of chlorine in the neutral NaAlCl<sub>4</sub> melt (about 10<sup>-6</sup> mole/cm<sup>3</sup> and 10<sup>-5</sup> cm<sup>2</sup>/s, respectively) confirm a low chlorine mass transfer rate to the carbon electrode. It was found that the addition of acetylene carbon black in the molten sodium improved the wetting of molten sodium on the surface of beta"-alumina tubes.

Different designs of the chlorine electrodes were tested and modeled theoretically. Limiting current density can be as high as 200 mA/cm<sup>2</sup> and an operating current density can be 180 mA/cm<sup>2</sup> at a -0.7-V chlorine electrode overpotential by using a bubble-through chlorine electrode.

A Na/beta"-alumina/NaAlCl<sub>4</sub>, Cl<sub>2</sub>/C circulating cell compared to the usual static cell design has indefinite capacity which depends on the size of the reservoirs. The theoretical specific energy is approached as the reservoirs

become large compared to the cell itself. The power is limited by the mass transfer rate of chlorine to the carbon electrode. It can be used as either a sodium-chlorine secondary battery or a sodium-chlorine fuel cell. Its measured open circuit voltage is between 3.78-4.13 V. After the problem of contact between molten sodium and beta"-alumina was partly solved, an output at 37.5 mA/cm<sup>2</sup> and 3.2 V was demonstrated.

Mathematical models provide a theoretical explanation for the performance of the Na/Cl<sub>2</sub> battery. The results of mathematical models match the experimental results very well. According to the result of the mathematical modeling, an output at 180 mA/cm<sup>2</sup> and 3.2 V can be obtained with optimized cell design.